

Interactive comment on “Stratospheric loss and atmospheric lifetimes of CFC-11 and CFC-12 derived from satellite observations” by K. Minschwaner et al.

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Authors' response to referee #2 comments for manuscript acp-2012-758 :

First, we wish to thank both reviewers for their time and effort in reviewing this paper. All comments and suggestions have been taken into consideration in revisions to the manuscript, and below we address specific points and questions raised by the reviewers.

In addition to the revisions suggested by both reviewers, there are a few additional changes to the analysis that we have implemented in order to produce improved lifetimes and uncertainties: 1. Means and uncertainties are calculated in $1/\tau$ space

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consistent with the fact that the loss rates, which appear in the denominator of the expressions for τ , are the primary quantities for our analysis. 2. CRISTA-1 and 2 results are averaged first, before computing the multi-instrument mean, since these two datasets are from the same instrument with similar retrievals. 3. Burdens have been recalculated based on a mean surface pressure of 985 hPa rather than 1000 hPa. 4. A typographical error found in the MIPAS CFC-11 steady state lifetime in table 1 has been corrected.

Referee #2 - Response to Specific Comments:

1. Referee comment: P. 28739, lines 4-6, in regards to using the SORCE solar flux data for one month (March 2004). Recent work has indicated possible problems with long term changes in the SORCE data (Lean and Deland, 2012, J. Climate, p. 2555; Swartz et al., 2012, ACP, p. 5937), although I assume this will not be an issue with using just the one month of data. However, will there be much of an impact on the computed lifetimes if instead the solar flux data corresponding to the specific times of the CFC measurements is used (eg, from the NRL solar spectral irradiance reconstruction - Lean, 2000)? For example, recent unpublished model calculations indicate that with everything else fixed, the CFC-11 lifetime changes by 1-2 years for solar max vs. solar min conditions using the Lean NRL solar flux (with a 2-3 year change for CFC-12). These changes are on the order of 2-3%, so this effect appears to be small, but the authors should at least add a sentence or two mentioning the potential impacts on the lifetimes of the solar flux variability

Authors' response: The issue of solar irradiance is an important one and we appreciate the reviewer pointing out that the manuscript lacked sufficient discussion. As noted, perhaps the most important effect is solar cycle variability. In the critical spectral region near 200 nm, the variation is estimated to be $\pm 4\%$ (Lean, 2000) although there is currently some controversy regarding solar cycle UV changes (e.g. Lean and Deland, 2012). The SORCE irradiances used in our work are from March 2004, a time period near the midpoint between solar maximum in 2000 and solar minimum in 2008.

Irradiances from this period may be considered as representative of mean solar conditions and therefore the most appropriate to use for calculating mean photolytic loss in steady-state lifetimes. We have increased the uncertainty in solar irradiance to 5% to allow for both photometric error and sub-sampling of the solar cycle, and have included a discussion of this issue in manuscript.

2. Referee comment: P. 28739, lines 22-25, “Additionally, there is a shift the peak in ozone mixing ratio.” This sentence has useful information, but as written is somewhat long and hard to follow. Please re-word/clarify, perhaps dividing into two sentences.

Authors’ response: Sentence has been revised by breaking into two.

3. Referee comment: P. 28742, lines 1-3, “ ...no corrections have been applied... ” – I don’t quite understand this sentence since lines 13-15 on the previous page states that the tropospheric mixing ratios have been adjusted to match the WMO, 2011 values. Please clarify. Also in regards to lines 10-11, “ ...MIPAS mixing ratios are larger by 10-20%...”, I assume this is mainly a measurement bias, and is not due to the 4% TD change in surface mixing ratio (WMO, 2011)? Please clarify.

Authors’ response: The manuscript was unclear on this. Tropospheric mixing ratios have been adjusted to match WMO 2011 values appropriate to for the time of each measurement. No corrections were applied to the data in Figure 3 to account for the changes in mixing ratio that occurred between the time of the two measurements. Thus one would expect, at minimum, that ACE would be lower than MIPAS by about 4% just from this difference alone. The reviewer is correct that most of the 10-20% difference is likely due to a measurement bias between the two datasets. The paper has been revised to clarify this.

4. Referee comment: p. 28743, lines 14-17: While the seasonal variation in the loss rates can be accounted for, some of the data sets have less than 1 year of coverage (e.g., 1 month for CRISTA-1, -2) so the seasonal variation in the constituent distributions will likely cause significant seasonal biases in the computed lifetimes. For exam-

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ple, unpublished model calculations (the same as noted above) indicate a seasonal variation of +/- 10 years for CFC-11 and +/-20 years for CFC-12 (due to the seasonal variations in both the constituent distributions and loss rates). There will also be a substantial QBO influence on the constituent distributions as well. Of course these are the limitations of the data sets and can't really be avoided, but the authors should include a statement or two noting that there are probable seasonal and interannual biases. Further to this point, I'm not sure if doing an equal-weighted mean lifetime in Table 1 is the best way to go. I would think giving the MIPAS and ACE data more weight (perhaps accounting for the number of months of data available) would be more appropriate for CFC-11.

Authors' response: The paper now includes a discussion of the impact of neglecting seasonal and interannual variations in CFC distributions. It is about a 5% effect based on the MIPAS dataset which has sufficient seasonal coverage in the tropics. Based on the ACE data, the interannual variability is a nearly 10% effect. Multi-instrument means have been converted to best-estimate lifetimes by averaging in $1/\tau$ in order to more accurately reflect the mean of the global loss calculations (which appear in the denominator of the lifetime expressions), which leads naturally to a recomputation of uncertainties in $1/\tau$ space (see comments to reviewer 1 as well).

5. Referee comment: P. 28745, lines 2-4, this is a bit confusing. Doesn't the smaller mixing ratio from CRISTA-2 (above 20 km) imply a shorter lifetime compared to MIPAS via eqn. 2? I would think the burden contribution to the older lifetime in CRISTA-2 vs. MIPAS is due to the larger burden in CRISTA-2 below 19km in Fig. 6? Please clarify this.

Authors' response: A smaller mixing ratio leads to a smaller calculated loss rate, via equation 4. Then from equation 2, a smaller loss rate leads to a longer lifetime. The global burden for CRISTA-2 is larger than for MIPAS primarily because of the larger tropospheric mixing ratios in 1997 compared with 2003.

6. Referee comment: Technical corrections, 1) p. 28736, line 5, change to: “ ...the assessment of CFC-11’s potential...” 2) p. 28740, for the leading factors in eqns. (3) and (4), change “2” to “4” since this should be the area of the earth (to get $5.1 \times 10^{18} \text{ cm}^2$). 3) Reference on p. 28753, line 10, change to “tropical” 4) Fig. 5, right panel: it would be good to include contours here since the point is made (p. 28743, lines 7-8) that the O(1D) loss contribution at 26-34 km (4-8%) is small but not zero. It’s very hard to discern this in the color-only figure as is.

Authors’ response: We have implemented suggestions 1,3, and 4, but not 2. These equations are based on a zonal average that has already been integrated in longitude, hence the factor of 2π . If one integrates over latitude then one obtains the correct value for the surface area of the earth.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 28733, 2012.

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